

Quantum insulating states of $F = 2$ cold atoms in optical lattices

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In this Letter we study various spin correlated insulating states of $F = 2$ cold atoms in optical lattices. We find that the effective spin exchange interaction due to virtual hopping contains an *octopole* coupling between two neighboring lattice sites. Depending on scattering lengths and numbers of particles per site the ground states are either rotationally invariant dimer or trimer Mott insulators or insulating states with various spin orders. Three spin ordered insulating phases are ferromagnetic, cyclic and nematic Mott insulators. We estimate the phase boundaries for states with different numbers of atoms per lattice site.

Recently, the scattering lengths of Rb atoms in the $F=2$ manifold in optical lattices have been studied experimentally [1]. These results, together with earlier theoretical estimates of scattering lengths [2] offer critical information about how atoms interact in the $F = 2$ manifold and are vital for the further understanding of their magnetic phases. Investigation of cold atoms with high spins in optical lattices not only improves our understanding of fundamental ideas in quantum magnetism, but might also lead to major breakthroughs in quantum information storing and processing [3, 4]. A few impressive theoretical efforts were already made to understand BECs of $F = 2$ sodium and rubidium atoms in optical traps [5, 6]. Depending on the two-particle scattering lengths in total spin $F = 0, 2, 4$ channels, the ground states of cold atoms can be either polar, cyclic or ferromagnetic condensates. The existence of various spin correlated states of cold atoms in optical lattices has also been suggested [7, 8].

In this Letter we shall address the following question: What are the possible insulating states of $F = 2$ cold atoms in optical lattices? We first summarize the main results of our investigation. a) When optical potentials are strong and exchange interactions are asymptotically weak, for certain numbers of particles per lattice site the ground states are rotationally invariant Mott insulators of dimers (i.e., singlets of two $F = 2$ atoms) or trimers (i.e., singlets of three $F = 2$ atoms). b) Decreasing the optical potential depth, depending on scattering lengths, results in various spin ordered insulating states such as cyclic, nematic and ferromagnetic insulators. c) The boundaries between different insulating phases are estimated (See Fig.(1)).

To obtain these results, we consider $F = 2$ cold atoms which have two-body scattering lengths, $a_{F=0,2,4}$, in three channels with total hyperfine spins $F = 0, 2, 4$. The two-body contact interaction is $\sum_{F=0,2,4} g_F \mathcal{P}_F$ where $g_F = 4\pi\hbar^2 a_F/M$ and \mathcal{P}_F are the projection operator which projects out total spin F states of two atoms. Alternatively this can be rewritten in terms of $a + b/2(\hat{F}^2 - 12) + 5c\mathcal{P}_0$ where \hat{F} is the total spin operator. Three interaction constants were calculated previously, $a = (4g_2 + 3g_4)/7$, $b = -(g_2 - g_4)/7$ and

$c = (g_0 - g_4)/5 - 2(g_2 - g_4)/7$ [5, 6]. To study the states in optical lattices, we find it is convenient to introduce a tensor representation $\psi_{\alpha\beta}^\dagger$, $\alpha, \beta = x, y, z$ for $F = 2$ states:

$$\begin{aligned}\psi_{xz}^\dagger &= \frac{1}{\sqrt{2}}(\psi_{-1}^\dagger - \psi_1^\dagger), & \psi_{yz}^\dagger &= \frac{i}{\sqrt{2}}(\psi_{-1}^\dagger + \psi_1^\dagger), \\ \psi_{xy}^\dagger &= \frac{i}{\sqrt{2}}(\psi_{-2}^\dagger - \psi_2^\dagger), & \psi_{zz}^\dagger &= \frac{2}{\sqrt{3}}\psi_0^\dagger, \\ \psi_{xx}^\dagger &= \frac{1}{\sqrt{2}}(\psi_{-2}^\dagger + \psi_2^\dagger) - \frac{1}{\sqrt{3}}\psi_0^\dagger \\ \psi_{yy}^\dagger &= \frac{-1}{\sqrt{2}}(\psi_{-2}^\dagger + \psi_2^\dagger) - \frac{1}{\sqrt{3}}\psi_0^\dagger\end{aligned}\quad (1)$$

where $\psi_{m_F}^\dagger$, $m_F = 0, \pm 1, \pm 2$ are the usual creation operators for $F = 2$ particles. The tensor operators $\psi_{\alpha\beta}^\dagger$ are symmetric and traceless, that is $\psi_{\alpha\beta}^\dagger = \psi_{\beta\alpha}^\dagger$ and $tr\psi^\dagger = 0$. We find this a convenient labelling of the five $F = 2$ states. The commutation relations are

$$[\psi_{\alpha\beta}, \psi_{\alpha'\beta'}^\dagger] = \delta_{\alpha\alpha'}\delta_{\beta\beta'} + \delta_{\alpha\beta'}\delta_{\beta\alpha'} - \frac{2}{3}\delta_{\alpha\beta}\delta_{\alpha'\beta'}. \quad (2)$$

The construction of rotationally invariant operators in this representation is straightforward. For instance, the number operator $\hat{\rho}$, the dimer or singlet pair creation operator \mathcal{D}^\dagger and the trimer or singlet of three atoms creation operator \mathcal{T}^\dagger are

$$\hat{\rho} = \frac{1}{2}tr\psi^\dagger\psi, \quad \mathcal{D}^\dagger = \frac{1}{\sqrt{40}}tr\psi^\dagger\psi^\dagger, \quad \mathcal{T}^\dagger = \frac{1}{\sqrt{140}}tr\psi^\dagger\psi^\dagger\psi^\dagger \quad (3)$$

where ψ^\dagger represents the tensor. The total spin operator \hat{F}_α , $\alpha = x, y, z$ is

$$\hat{F}_\alpha = -i\epsilon_{\alpha\beta\gamma}\psi_{\beta\eta}^\dagger\psi_{\eta\gamma}, \quad [\hat{F}_\alpha, \hat{F}_\beta] = i\epsilon_{\alpha\beta\gamma}\hat{F}_\gamma. \quad (4)$$

Finally, \hat{F}_α commutes with rotationally invariant operators such as \mathcal{D}^\dagger and \mathcal{T}^\dagger ; generally one finds that $[\hat{F}_\alpha, tr(\psi^\dagger)^n] = 0$. For $F = 2$ cold atoms in optical lattices, we employ the following Hamiltonian

$$\mathcal{H} = \frac{a_L}{2} \sum_k (\hat{\rho}_k^2 - \hat{\rho}_k) + \frac{b_L}{2} \sum_k (\hat{F}_k^2 - 6\hat{\rho}_k) + 5c_L \sum_k \mathcal{D}_k^\dagger \mathcal{D}_k - t \sum_{\langle kl \rangle} (\psi_{k,\alpha\beta}^\dagger \psi_{l,\beta\alpha} + h.c.). \quad (5)$$

Here k is a lattice site index and $\langle kl \rangle$ are neighboring sites. t is the one-particle hopping amplitude. a_L, b_L, c_L are, respectively, proportional to a, b, c introduced earlier; $a_L(b_L, c_L) = a(b, c) \int d\mathbf{r} (\psi_0^* \psi_0)^2$ and ψ_0 is the on-site local ground state wavefunction. $\mathcal{D}^\dagger \mathcal{D}$ plays the role of projection operator \mathcal{P}_0 discussed before. For most cold atoms, the absolute values of b_L, c_L are much smaller than a_L and we work in this limit unless specified. When the optical potential depth is small and t is much larger than a_L , cold atoms effectively are weakly interacting and condense. The results obtained using Eq.(5) are identical to the ones previously obtained and we do not present them in this Letter. Here we focus on the physics of insulating states when t is much smaller than a_L .

The effective low energy Hamiltonian for a Mott insulator with $M(> 1)$ atoms per lattice site is

$$\begin{aligned} \mathcal{H} &= \frac{b_L}{2} \sum_k (\hat{F}_k^2 - 6\hat{\rho}_k) + 5c_L \sum_k \mathcal{D}_k^\dagger \mathcal{D}_k \\ &- J_0 \sum_{\langle kl \rangle} (Q_{k;\alpha\beta,\alpha'\beta'}^\dagger Q_{l;\alpha\beta,\alpha'\beta'} + h.c.); \\ Q_{k;\alpha\beta,\alpha'\beta'}^\dagger &= \psi_{k,\alpha\beta}^\dagger \psi_{k,\alpha'\beta'} - \\ &\frac{1}{10} \text{tr} \psi_k^\dagger \psi_k (\delta_{\alpha'\alpha} \delta_{\beta\beta'} + \delta_{\alpha\beta'} \delta_{\beta\alpha'} - \frac{2}{3} \delta_{\alpha\beta} \delta_{\alpha'\beta'}) \end{aligned} \quad (6)$$

where $J_0 = t^2/a_L$. The virtual hopping between two neighboring sites results in an exchange interaction of the form of *octopole* coupling as shown above. This is valid for all Mott states with the number of particles M larger than one. An exchange interaction of quadrupole type has been derived recently for $F = 1$ cold atoms in optical lattices [7, 9, 10].

The spin-ordered Mott insulating states under consideration (with the crystal translational symmetry) are

$$|\chi\rangle = \prod_k \frac{(\psi_k^\dagger \chi)^M}{\sqrt{M!}} |0\rangle, \text{tr} \chi \chi^* = \frac{1}{2}; \quad (7)$$

χ is a traceless symmetric complex tensor and M is the number of atoms per site. We calculate the χ -dependent part of the mean field energy per particle,

$$\begin{aligned} E_{MI} &= (M-1)A(\chi, \chi^*) - \frac{16}{5} J_{ex} M; \\ A(\chi, \chi^*) &= 4b_L \text{Tr}[\chi, \chi^*]^2 + 2c_L \text{tr} \chi^2 \text{Tr} \chi^{*2} \end{aligned} \quad (8)$$

Here $J_{ex} = zJ_0$, z is the number of neighboring lattice sites. Minimization of this energy subject to the constraint of the normalization of χ in Eq.(7) can be carried

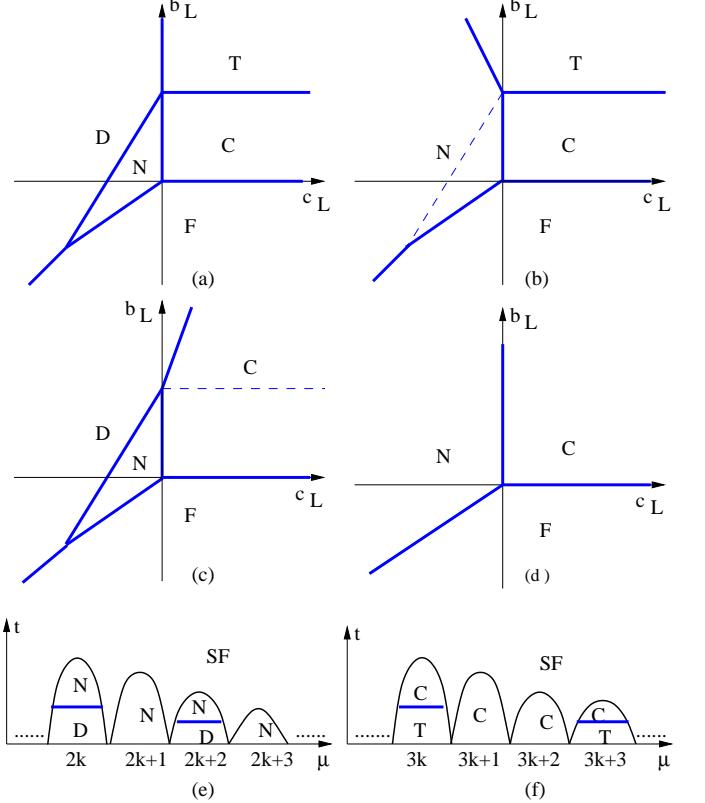


FIG. 1: (Color online) Schematic of phase diagrams for spin correlated Mott states of $F = 2$ cold atoms in optical lattices. In (a)-(d), the phase diagrams are plotted in a plane of two interaction constants b_L, c_L for different M (the number of particles per lattice site). Only insulating states are shown. a_L, b_L are defined in terms of scattering lengths after Eq. (5). In (e) and (f), the vertical axis is the hopping t and the horizontal axis is the chemical potential μ ; the numbers along the μ -axis indicate the number of particles per lattice site in Mott states (states inside lobes). Outside the lobes are superfluids which we do not discuss in this article. (a) for $M = 3k = 2k'$; (b) for $M = 3k \neq 2k'$ (c) for $M = 2k' \neq 3k$; (d) for $M \neq 3k \neq 2k'$ or for $M = 1$. (e) for $-c_L \gg b_L > 0$; (f) for $c_L \gg b_L > 0$. N,C,F, D, T stand, respectively, for nematic, cyclic, ferromagnetic, dimer and trimer Mott insulating states; In (b) and (c), there are *no* phase transitions across dashed lines (guide for eyes only). Phase boundaries (thick blue lines) are estimated by applying Eq. (8) and comparing the energies obtained in Eq. (12-18).

out using the method of the Lagrangian multiplier. We provide the results here:

1) $b_L > 0$ and $c_L > 0$. The traceless tensor χ satisfies

$$\text{tr} \chi^2 = [\chi, \chi^*] = 0. \quad (9)$$

There are two discrete *root* solutions $\chi_{R\pm}$ which have only diagonal components, $\chi_{xx} = 1/\sqrt{6}$, $\chi_{yy} = \exp(\pm i2\pi/3)/\sqrt{6}$ and $\chi_{zz} = \exp(\pm i4\pi/3)/\sqrt{6}$ and $\chi_{\alpha\beta} = 0$ when $\alpha \neq \beta$. They are also invariant under a cyclic subgroup C_3 of the $\text{SO}(3)$ rotation group. The full set

of solutions can be obtained by applying SO(3) rotation, and $U(1)$ gauge transformation. These solutions correspond to cyclic insulating states.

2) $b_L > c_L/4$ and $c_L < 0$. Up to a phase, χ satisfies

$$\text{tr}\chi^2 = \frac{1}{2}, \chi = \chi^*. \quad (10)$$

For diagonal matrices which satisfy the above conditions, the matrix elements have to fall on an elliptical curve; that is, $\chi_{xx}^2 + \chi_{xx}\chi_{yy} + \chi_{yy}^2 = 1/4$, $\text{tr}\chi = 0$ and $\chi_{\alpha\beta} = 0$ if $\alpha \neq \beta$. The full set of solutions can be obtained by applying $SO(3)$ and $U(1)$ rotation. These states are characterized by *traceless real symmetric* tensors (up to a phase) and $\langle \hat{F}_\alpha \rangle = 0$, $\alpha = x, y, z$; moreover, the corresponding expectation value of the nematic order operator $\psi_{\alpha\gamma}^\dagger\psi_{\gamma\beta} - 1/3\delta_{\alpha\beta}\text{tr}\psi^\dagger\psi$ is nonzero. They represent nematic insulating states (see more discussions in Ref.10).

3) $b_L < 0$ and $b_L < c_L/4$. χ satisfies

$$\text{tr}\chi^2 = 0, \chi^*\chi\chi^* - \chi^*\chi^*\chi - \text{tr}[\chi, \chi^*]^2\chi^* = 0 \quad (11)$$

and $[\chi, \chi^*] \neq 0$. An example of solutions is $\chi_{z\alpha} = 0$ ($\alpha = x, y, z$) and $\chi_{xx} = -\chi_{yy} = -i\chi_{xy} = 1/\sqrt{8}$. This corresponds to a ferromagnetic insulating phase.

So three spin ordered Mott insulating states are ferromagnetic, nematic and cyclic states (indicated by subscripts F, N, C respectively); the corresponding energies per atom are

$$\begin{aligned} E_F &= 2(M-1)b_L - \frac{16}{5}MJ_{ex} \\ E_N &= (M-1)\frac{c_L}{2} - \frac{16}{5}MJ_{ex}, E_C = -\frac{16}{5}MJ_{ex} \end{aligned} \quad (12)$$

In addition, one should also take into account rotationally invariant insulating states. The physics of these states depends on the numbers of particles per lattice site and below we discuss different situations separately.

i) $M = 2k = 3k'$ (k, k' are integers). When b_L and $-c_L$ are positive and J_{ex} approaches zero, the Hamiltonian has the lowest energy eigenstate which is rotationally invariant and has a maximal number of singlet pairs, or *dimers*. It has the following wavefunction

$$|D\rangle = \prod_k \left(\frac{\text{tr}\psi_k^\dagger\psi_k^\dagger}{\sqrt{40}} \right)^{\frac{M}{2}} |0\rangle, E_D = (M+3)\frac{c_L}{2} - 3b_L \quad (13)$$

The state $|D\rangle$ (up to a normalization factor) is an insulator of singlet pairs or dimers; following Eq.(12) and Eq.(13), it has a lower energy than the nematic state when the exchange interaction is small, i.e.,

$$J_{ex} < J_D^c, J_D^c = \frac{5}{16M}(3b_L - 2c_L). \quad (14)$$

In dimer Mott states, all atoms are paired in singlets and $\langle D|\mathcal{D}^\dagger\mathcal{D}|D\rangle = M(M+3)/10$.

When b_L and c_L are both positive and the exchange coupling is zero, the ground state of the Hamiltonian is still an M-atom spin singlet but has to be *annihilated* by the dimer operator \mathcal{D} . For $M = 3$, such a state can be created by the trimer creation operator \mathcal{T} ; the wavefunction and its energy are

$$|T\rangle = \prod_k \frac{1}{\sqrt{140}} \text{tr}(\psi_k^\dagger)^3 |0\rangle, E_T = -3b_L \quad (15)$$

Note that $\mathcal{D}|T\rangle = 0$ and $\langle T|\mathcal{D}^\dagger\mathcal{D}|T\rangle = 0$.

For more than three particles, one can show that singlet states of the form $\text{tr}\psi^n$ with $n > 3$ can always be rewritten in terms of two *fundamental* singlet creation operators: $\text{tr}\psi^2$ for a dimer and $\text{tr}\psi^3$ for a trimer. Generally, singlet states for $M = 2k_1 + 3k_2$ atoms can be expressed in terms of $(\text{tr}\psi^\dagger\psi^\dagger)^{k_1}$ $(\text{tr}\psi^\dagger\psi^\dagger\psi^\dagger)^{k_2}$. For instance, direct calculations show that $\text{tr}(\psi^\dagger)^4|0\rangle$ is identical to $(\text{tr}\psi^\dagger\psi^\dagger)^2|0\rangle$ and $\text{tr}(\psi^\dagger)^5|0\rangle$ is identical to $\text{tr}(\psi^\dagger)^2\text{tr}(\psi^\dagger)^3|0\rangle$. To construct an M-particle singlet without singlet pairs, the only candidate to consider is $(\text{tr}\psi^\dagger\psi^\dagger\psi^\dagger)^k|0\rangle$ since all other states explicitly involve $(\text{tr}\psi^\dagger\psi^\dagger)^n$, the dimer creation operator. Therefore, for an arbitrary M, a 3k-atom singlet *without dimers* can be written as $|T\rangle = \prod_k \mathcal{P}_T(k)(\text{tr}(\psi^\dagger)^3)^k$; (up to the normalization factor) here $\mathcal{P}_T(k)$ is a projection operator which further projects out states without dimers at site k , i.e. $\mathcal{D}_k\mathcal{P}_T(k) = 0$. Taking into account that $\hat{F}_k^2|T\rangle = \mathcal{D}_k|T\rangle = 0$, we find that the energy per atom for a trimer state is independent of M , $E_T = -3b_L$. It has a lower energy than the cyclic state when

$$J_{ex} < J_T^c, J_T^c = \frac{15}{16M}b_L. \quad (16)$$

By further comparing $E_{D,T}$ and $E_{C,N,F}$ we obtain the mean field phase diagram; we find the mean field phase boundaries between rotationally invariant trimer and dimer states and three spin ordered phases, taking into account Eq.(12), Eq.(13) and Eq.(15). The results are summarized in Fig.(1 a). The boundary between T - and D -phase is $c_L = 0$, C - and F -phase is $b_L = 0$, T and C -phase is $b_L = 16MJ_{ex}/15$, between D - and N -phase is $b_L = 2c_L/3 + 16MJ_{ex}/15$, between N - and F -phase is $b_L = c_L/4$, and finally between D and F -phase is $b_L = (M+3)c_L/(4M+2) + 16MJ_{ex}/5(2M+1)$.

ii) $M = 3k = 2k' + 1$. Following the general construction outlined in i), the trimer states are ground states in optical lattices when c_L and b_L are both positive and J_{ex} is approaching zero. However, when c_L is negative, b_L is positive, as J_{ex} approaches zero, at each lattice site k' singlet pairs or dimers are formed leaving the last atom unpaired. So the system effectively is a lattice with an $S = 2$ spin at each lattice site. The situation is similar to what happens to $F = 1$ atoms in optical lattices with

an odd number of atoms per site [10]. Following the discussions on the case for one atom per lattice ($M = 1$) below, we find that the ground state remains to be nematically ordered down to $J_{ex} = 0$ as far as c_L is negative and b_L is positive. Only four phases (trimer Mott insulators and spin ordered cyclic, ferromagnetic, nematic Mott states) appear in the phase diagram as shown in Fig.(1 b). The phase boundary between T - and N -phase is $b_L = (1 - M)c_L/6 + 16M J_{ex}/15$ which approaches $c_L = 0$ as M becomes infinity. The other phase boundaries are the same as in Fig. (1 b) discussed before.

iii) $M = 2k = 3k' \pm 1$. When c_L is negative and b_L is positive and the exchange interactions are small, the ground states are dimer insulating states. When c_L and b_L are positive and J_{ex} approaches zero, at each lattice site k' trimers are formed; the last atom (for $M = 3k' + 1$) or two (for $M = 3k' + 2$) which do not participate in the trimer formation are in an $F = 2$ spin collective state. Effectively at each site only the $F = 2$ state is relevant for spin ordering and the system is again equivalent to a lattice spin Model for $S = 2$ spins. This limit is equivalent to $M = 1$ case studied below and we find that the cyclic state remains to be the ground state as J_{ex} becomes zero, provided that b_L and c_L are positive. Four phases that appear in the phase diagram are dimer Mott insulators and spin ordered cyclic, ferromagnetic, nematic Mott states, as shown in Fig.(1 c). The boundary between D and C is $b_L = 16M J_{ex}/15 + (M+3)/6c_L$; again it approaches $c_L = 0$ as M becomes infinity. The other boundaries are identical to those in Fig.(1 a).

iv) Finally, we would like to address the special case of $M = 1$. The effective Hamiltonian derived before is only valid when $M > 1$. Here we directly calculate the exchange interaction between two adjacent $F = 2$ spins due to virtual hopping and obtain the following Hamiltonian

$$\frac{\mathcal{H}}{J_0} = - \sum_{\langle kl \rangle} \sum_{F=0,2,4} \frac{\tilde{a}}{a_F} \mathcal{P}_F(kl) \quad (17)$$

Here $\mathcal{P}_F(kl)$ projects out a total spin F state of two atoms on neighboring sites $\langle kl \rangle$. \tilde{a} is defined as $\tilde{a} = (4a_2 + 3a_4)/7$. For two adjacent atoms with total spin $F = 1, 3$, the exchange interaction vanishes. We calculate the energy per particle using the wavefunction specified by Eq.(7) with $M = 1$ and derive an expression for the energy as a function of χ . The χ -dependence of the energy per atom can be shown identical to that in Eq.(8). We again obtain three phases and the energies per atom of cyclic, nematic and ferromagnetic states are

$$\begin{aligned} \frac{E_F}{J_0} &= -\frac{\tilde{a}}{a_4}, \quad \frac{E_c}{J_0} = -\frac{3\tilde{a}}{7a_4} - \frac{4\tilde{a}}{7a_2}, \\ \frac{E_N}{J_0} &= -\frac{18\tilde{a}}{35a_4} - \frac{2\tilde{a}}{7a_2} - \frac{\tilde{a}}{5a_0}. \end{aligned} \quad (18)$$

When the difference between scattering lengths is small, one can easily verify that the phase boundaries of these three insulating states are identical to those of three condensates discussed previously. The results are summarized in Fig.(1 d). The boundaries between F and C , C and N and N and F are, respectively, $b_L = 0$, $c_L = 0$ and $b_L = c_L/4$ [11]. Finally we present the results in terms of hopping t and chemical potential μ of $F = 2$ atoms in two parameter regions: 1) $-c_L \gg b_L > 0$ (Fig.(1e)); and 2) $c_L \gg b_L > 0$ (Fig.(1f)); the spin ordering exhibits mod 2 (Fig.(1e)) and mod 3 (Fig.1f) behaviors.

In conclusion, we find that $F = 2$ cold atoms in optical lattices in addition to forming spin ordered Mott states, can also be in states that only consist of dimers or trimers. These results are relevant to Rb and Na atoms in optical lattices. According to earlier estimates[2], ^{87}Rb atoms fall nearly on the $c_L = 0$ line ($b_L > 0$), a phase boundary between the cyclic and nematic insulating phases. For ^{23}Na , $b_L = -4.61c_L > 0$; the insulating state of ^{23}Na atoms is nematic when J_{ex} is large; for $M = 2k$ as the optical potential decreases, a quantum phase transition occurs and the ground state becomes a dimer Mott state. However, the error bars in estimates of scattering lengths might lead to uncertainties in determining ground states. Finally, dimer or trimer Mott states have fully gapped spin excitations and are robust; spin ordered states in $3D$ optical lattices are stable against quantum fluctuations (see similar discussions in Ref.[10]). In a finite trap because of coexistence of Mott states with different occupation numbers[12], the spin order spatially alternates between dimer (trimer) and nematic (trimer) ones. This work is supported by the office of the Dean of Science, UBC, and NSERC, Canada. FZ currently is an A. P. Sloan fellow.

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